

ISOTOPIC FRACTIONATION AS A PROBE OF EVAPORATION PROCESSES IN STELLAR ENVIRONMENTS: Hiroko Nagahara and Kazuhito Ozawa, Geol. Inst., Univ. Tokyo, Hongo, Tokyo 113, Japan.

Development of isotopic fractionation during evaporation in a solid or liquid droplet was modeled taking the effect of elemental diffusion in a condensed phase into consideration. The mode of isotopic fractionation is divided into “Rayleigh fractionation”, “zoned fractionation”, or “no fractionation” regime; in the Rayleigh fractionation regime evaporation is rate limiting, in the zoned regime both diffusion and evaporation are rate limiting, and in the no fractionation regime diffusion is rate limiting. The results are applied to show the three regimes in the space of initial grain size and temperature for forsterite, SiO₂, and MgO-SiO₂ melt. Magnesium in forsterite and O in silicate melt can possess records of high temperature. The results predict that we can know whether minerals, chondrules or CAIs suffered evaporation if isotopic zoning profile in them are measured.

Evaporation is one of the most dominant and important time-dependent processes in stellar environments. Although Rayleigh fractionation is often assumed for evaporation, the situation is achieved only in the case when the inside of the solid or liquid is homogenized instantaneously. The role of diffusion in isotopic fractionation was stressed by [1] and modelled by [2]. Because we are interested in three-dimensional distribution of isotopes in a solid experienced extensive evaporation, we have developed a spherical model which has a moving boundary. By solving the equation, we have investigated the development of isotopic compositions as a function of time and size of the sphere.

The nondimensionalized diffusion equation is $\frac{\partial x}{\partial t} = \frac{\partial^2 x}{\partial r^2} + \frac{2}{r} \frac{\partial x}{\partial r}$, where x is the abundance ratio of i th isotope against the dominant isotope, t is time, and r is radial distance from the center of a sphere. Boundary conditions are $\left(\frac{\partial x}{\partial r}\right)_{r=0} = 0$ and $\left(\frac{\partial x}{\partial r}\right)_{r=R} = \beta(1-\alpha)x$, where α is the fractionation factor and $\beta = \frac{R_0 J^i}{D_i}$ is a dimensionless parameter. J^i is the linear evaporation rate of the substance, R_0 is the initial radius of the sphere, and D_i is the diffusion coefficient of the i th component. The dimensionless parameter represents the ratio of time scale of diffusion against that of evaporation. We have assumed a constant evaporation rate, a constant isotopic fractionation factor, and constant major isotope concentration.

Distribution of isotopic composition in an evaporating spherical grain varies drastically with β . When β is small (<0.1), a grain is isotopically homogeneous and becomes heavier with time (Fig. 1a). This is the situation where elemental diffusion in the grain is rapid enough and it corresponds to the case that Rayleigh fractionation is realized. Degree of isotopic fractionation in this case is largest. When β is intermediate ($1.0 < \beta < \text{several tens}$), a grain is isotopically zoned, which becomes heavier with time (Fig. 1b). This is the situation where elemental diffusion and evaporation are competing. Degree of isotopic fractionation becomes smaller with increasing β . When β is larger than several tens, a grain is almost homogeneous with steep compositional zoning near the very surface (Fig. 1c). Isotopic composition of the grain does not change with time. In this case, elemental diffusion in the grain is slow enough not to change the isotopic composition in the grain and therefore only evaporation from the surface is observed. The value of fractionation factor does not largely change above tendencies but changes the degree of isotopic fractionation. Degree of fractionation and the compositional difference from the center to the rim of a grain become larger with decreasing fractionation factor.

The mode of isotopic fractionation can be divided into three regimes on the basis of above results: “Rayleigh fractionation regime” where evaporation is the limiting process ($\beta < 1$), “zoned fractionation regime” where both evaporation and diffusion are limiting processes ($1 < \beta < 100$), and “no fractionation regime” where diffusion is the limiting process ($\beta > 100$). By inserting elemental diffusion rates and evaporation rates, isotopic fractionation mode was investigated in the grain size-temperature space for forsterite, SiO₂ and SiO₂-MgO melt. The data source for evaporation are [3] for forsterite, [4] for SiO₂, and [5] for MgO-SiO₂ melt, and that for diffusion rate [6] for Mg, Si, and O in forsterite, [7] for Si and O for SiO₂, and [8] Mg and O for melt. The results for forsterite is shown in Fig. 2. Because the evaporation rate in the forsterite-H₂ system varies with P/H₂ [3], the conditions for three isotopic fractionation regimes vary with total pressure. The “Rayleigh” or “zoned” regimes are expected for grains smaller than 1mm in a wide temperature range. The results predict that Mg in forsterite and O in melted objects such as chondrules and CAIs are candidates for marker of high temperature events. Absence of isotopic fractionation does not mean that the object had not suffered an evaporating event, which supports the discussion by [9] against [10].

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References: [1] Wang, J. et al. (1991) LPSC XXII,1461 [2] Wang, J. et al. (1994) ICOG-USGS circular [3] Nagahara, H. & Ozawa, K. (1996) GCA 60,1445 [4] Nagahara, H. (unpub. data) [5] Nagahara, H. and Ozawa, K. (1996) LPSC XXVII,927 [6] Morioka, M.(1991) in Advances in Physical Geochemistry 8 176 [7] Schactner, R. & Sockel, H.G. (1977) CMP76,440 [8] Dunn, T. (1983) GCA47 1923 [9] Esat, T. (1996) GCA 60,3755 [10] Humayun & Clayton (1995) GCA59 2131.

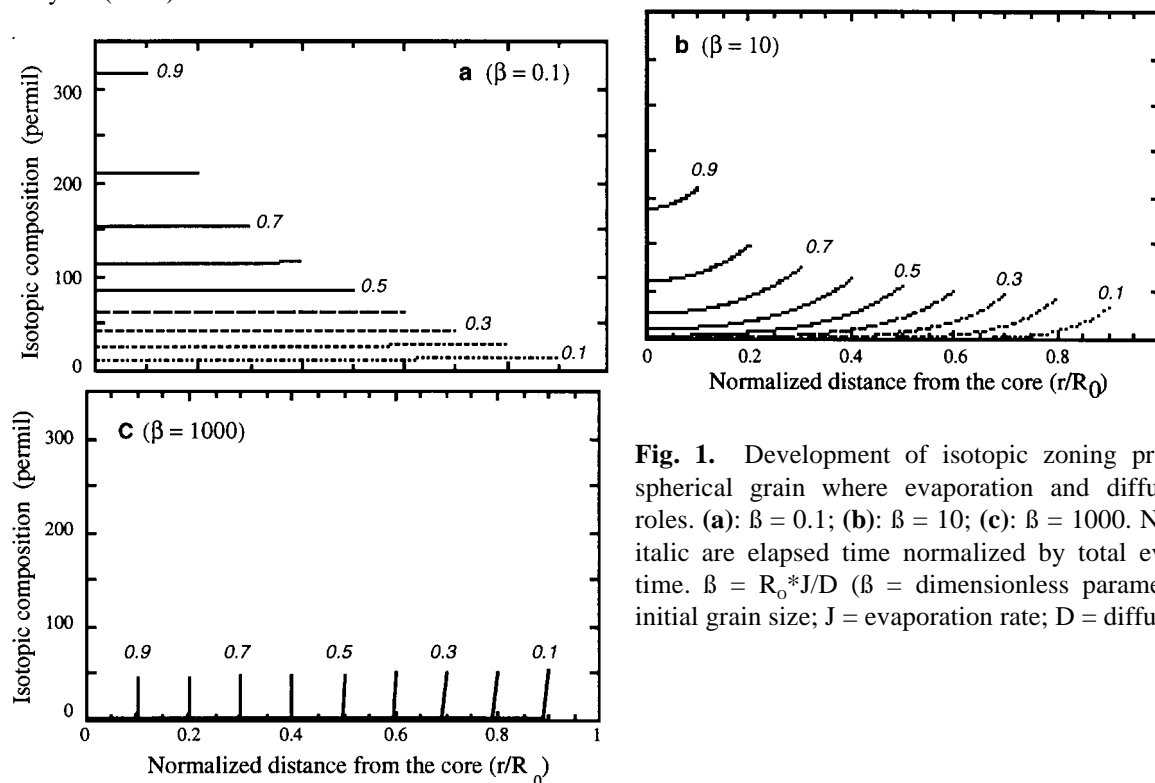


Fig. 1. Development of isotopic zoning profile in a spherical grain where evaporation and diffusion play roles. (a): $\beta = 0.1$; (b): $\beta = 10$; (c): $\beta = 1000$. Numbers in italic are elapsed time normalized by total evaporation time. $\beta = R_0 \cdot J/D$ (β = dimensionless parameter; R_0 = initial grain size; J = evaporation rate; D = diffusion rate).

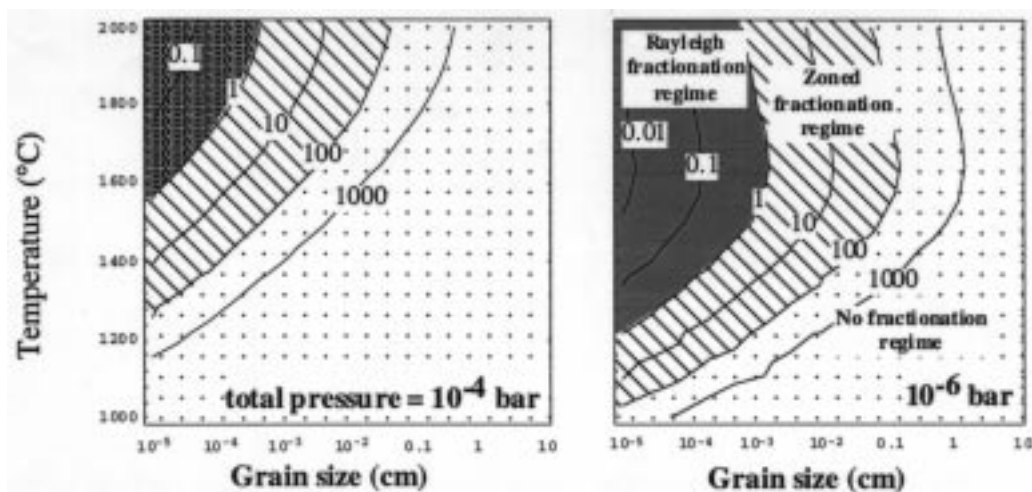


Fig. 2. Three isotopic fractionation regimes in the space of grain size and temperature for forsterite in the Mg_2SiO_4 - H_2 system. Numbers on curves are β values.